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ORIGINAL PAPER

A Study of the Magnetic Properties of $Bi_{1.64-x}Pb_{0.36}Cd_xSr_2Ca_2Cu_3O_y$ Superconductor

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Abstract The flux pinning energy and magnetic properties of $Bi_{1.64-x}Pb_{0.36}Cd_xSr_2Ca_2Cu_3O_y$ (BPCSCCO) with $x = 0.0$, 0.02, 0.04 and 0.06 were studied. A series of Bi-2223 superconductor samples with a nominal composition of BPCSCCO was synthesized and the effect of Cd substitution for Bi was investigated. As a result, Cd addition has been found to improve the superconducting properties of the Bi-Pb-Sr-Ca-Cu-O system. The effects of the annealing time and the amount of Cd doping on the structure, AC magnetic susceptibility, ρ–T curves and flux pinning energy were investigated. Also, for all samples the relation between the current and voltage in the mixed state was found to follow the model relationship $V = \alpha I^{\beta}$. The maximum value of β is 22.30, which is obtained for the sample with an annealing time of 270 h and a Cd content of 0.04.

Keywords ρ–T curves · Flux pinning energy · Magnetic susceptibility

1 Introduction

So far, more than two decades have passed since the discovery of Bi-based superconductors and many investigations of the substitution of various elements such as Sm [1], Sn and Sb [2, 3], Ag [4, 5], Cd [6–8] and Zr, Hf, Mo and W [9] into Bi–Sr–Ca–Cu–O have been reported, but it seems that the effect of substitution of cadmium for Bi on superconductors of the Bi-2223 phase has not been studied. Since the discovery of high-T_c superconductivity in the Bi–Sr–Ca–Cu–O system, three superconducting phases of $(Bi,Pb)_2Sr_2Ca_{n-1}Cu_nO_y$ have been identified [10]. They include the Bi-2201 phase ($n = 1$, $T_c \approx 20$ K), the Bi-2212 phase ($n = 2$, $T_c \approx 85$ K) and the Bi-2223 phase ($n = 3$, $T_c \approx 110$ K). Here, we have used the abbreviations Bi-2201, Bi-2212 and Bi-2223 phases for $n = 1, 2$ and 3, respectively. However, to obtain the pure Bi-2223 phase is very difficult and its formation is influenced by many preparation conditions such as composition, annealing time and temperature, atmosphere and pressure during sintering, the type and quantity of the dopant, heat-treatment method, and operational procedures. Numerous studies in doping have been made in order to synthesize and stabilize the Bi-2223 phase in the Bi–Sr–Ca–Cu–O system and to raise T_c [11–13]. In order to study the doping effect on T_c and the formation of Bi-2223 phase, the high T_c phase development must be studied under specified conditions. Takano et al. [14] and others showed that the incorporation of Pb in the nominal Bi–Sr–Ca–Cu–O composition of this system is very effective in increasing the volume fraction of the high-T_c phase [15–17]. In power applications, it is necessary to fabricate superconducting materials with high critical current densities J_c. It has been realized that polycrystalline superconductors can be described as arrays of superconducting grains weakly coupled by Josephson junctions. These weakly coupled grains are known to limit the J_c values of superconductors [18, 19]. A tremendous effort has been applied to improve the links between the grains and the properties of the Bi-based superconductor. The effect of Cd substitution for Bi in Bi-2201 phase of the Bi–La–Ca–Cu–O system [7] has been investigated; the results of the investigations show

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that the substitution of Cd for Bi does not affect T_c and also the oxygen content decreases monotonically with addition of Cd content. The differences between our work and the paper mentioned above are as follows. First of all, the Bi-based superconductor in our work is Bi-2223 phase. Second, our results show that the Cd substitution for Bi has increased the transition temperature, flux pinning energy and Bi-2223 phase percentage.

2 Materials and Methods

Samples of $Bi_{1.64-x}Pb_{0.36}Cd_xSr_2Ca_2Cu_3O_y$ with $x = 0.0$, 0.02, 0.04 and 0.06 were prepared by a solid state reaction method. Fine powders of Bi_2O_3, PbO, $SrCO_3$, $CaCO_3$, CuO and CdO (purity $\geq 99.9\%$) were used. In the first part of our experiments, $Bi_{1.6}Pb_xCd_zSr_2Ca_2Cu_3O_y$, where $(z, x) = (0.01, 0.39)$, (0.02, 0.38), (0.03, 0.37), (0.04, 0.36), (0.05, 0.35), (0.06, 0.34), (0.07, 0.33), (0.08, 0.32), (0.09, 0.31) and (0.1, 0.3) with different periods of sintering time (90, 180 and 270 h), were synthesized. The results showed that the optimum condition happened at $z = 0.04$ and $x = 0.36$, with the sintering time of 270 h. Considering this, next we studied the effect of doping in the vicinity of the optimum conditions. So, we synthesized $Bi_{1.64-x}Pb_{0.36}Cd_xSr_2Ca_2Cu_3O_y$ (BPCSCCO) with $x =$ 0.0, 0.02, 0.04 and 0.06. The required quantities of reagents were weighed ($\Delta m = 10^{-4}$ mg) and mixed. In order to prevent the growth of additional phases during the process, the powders were ground and milled for 1 h. The mixed powders were calcined at 820 °C for 15 h in air. Then they were taken out from the furnace, re-ground and pressed (with 250 bar) into the shape of pellets and bars. Then, they were put in an alumina crucible and placed in the furnace and the procedure of sintering was carried out in an air atmosphere. All of the samples were given the Meissner effect test in liquid nitrogen. The ρ–T curves (at a constant direct current of 40 mA) and the flux pinning energy of the samples were measured by the standard four-probe method. The AC magnetic susceptibility measurements were performed using a Lake Shore AC susceptometer, model 7000. The X-ray diffraction (XRD) patterns of the samples were taken with a Philips X-ray diffractometer, model PW1840.

3 Results and Discussion

The XRD measurements of the samples are shown in Fig. 1. Based on the XRD measurements, it is observed that by substitution of Cd instead of Bi up to an amount of 0.04, the Bi-2223 phase increases in the samples. From the XRD results, one will notice that the volume of Bi-2223 phase decreases on increasing the amount of cadmium (sample d). The results of XRD show that the optimum amount of cadmium is about $x = 0.04$ (sample c), which has the highest volume fraction of Bi-2223 phase. The volume fraction of the phases can be estimated by various methods [20]. The volume fractions of the phases for all the samples are given in Table 1.

Figure 2 shows that on increasing $x = 0.0$ to 0.04, T_c (onset) and T_c (offset) of the samples will increase. When the amount of cadmium is more than 0.04, T_c (onset) and T_c (offset) of the samples decrease.

Figure 3 shows the ρ–T curves of the sample with $x = 0.04$ at different annealing times. One can observe that on increasing the annealing time (t) up to 270 h, T_c (onset) and T_c (offset) of the samples will increase. The results of measurements show that the maximum T_c (onset) and T_c (offset) were found for the sample of

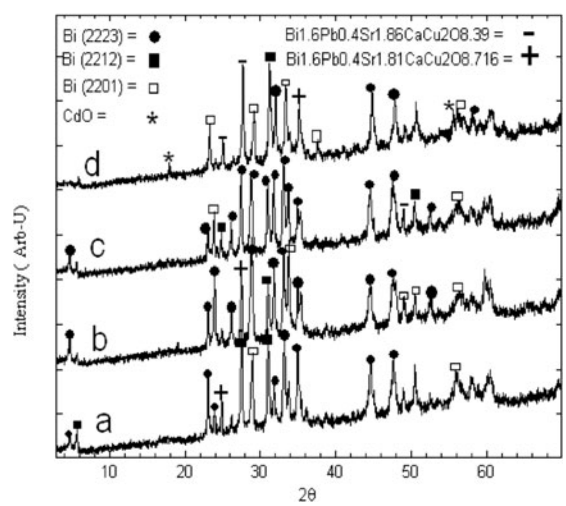

Fig. 1 XRD patterns of samples with different amounts of cadmium, 0.0 (*a*), 0.02 (*b*), 0.04 (*c*) and 0.06 (*d*), and an annealing time of 270 h

Table 1 Relative volume fractions of Bi-2223, Bi-2212, Bi-2201, Bi-21.8612, CdO and Bi-21.8112 with an annealing time of 270 h

Sample	*a*	*b*	*c*	*d*
Amount of Cd	0.0	0.02	0.04	0.06
Bi-2223 (%)	48.37	63.37	77.50	25.48
Bi-2212 (%)	31.57	7.19	3.27	11.36
Bi-21.8612 (%)	–	6.37	4.05	15.41
Bi-21.8112 (%)	5.20	–	–	7.18
Bi-2201 (%)	14.86	23.07	15.18	31.29
CdO (%)	–	–	–	9.28

$Bi_{1.6}Pb_{0.36}Cd_{0.04}Sr_2Ca_2Cu_3O_y$ which was annealed for 270 h. We attribute the increase of T_c (offset) to enhancement of the Bi-2223 phase in the sample. Also, on increasing the Cd content to more than 0.04, the amounts of the other phases in the Bi-Sr-Ca-Cu-O system will increase, which causes a decrease in T_c. The resistivity vs. temperature curves of all the samples are measured in the range of 77–300 K (see Fig. 4); however in this paper resistivity values are reported up to 200 K.

Figure 5 shows the results of magnetic susceptibility measurements for samples with amounts of $x = 0.0$, 0.04 and 0.06. In particular, the imaginary component, χ'', of the AC magnetic susceptibility has been widely used to probe the nature of weak links in polycrystalline superconductors. They are also employed to estimate some of the important physical properties, such as the critical current density and the effective volume fraction of the superconducting grains [21]. The real part of the AC magnetic susceptibility, χ', in polycrystalline samples shows two drops as the temperature is lowered before the onset of the diamagnetic transition and correspondingly the derivative of $\chi'(T)$ displays two peaks. The first sharp drop at the critical temperature is due to the transition within the grains and the second gradual change is due to the occurrence of the superconducting coupling between grains. The imaginary part, χ'', shows a peak which is a measure of the dissipation in the sample. When the peak of χ'' shifts to lower temperatures and broadens, the intergranular coupling between the grains, the critical current density and the flux pinning energy decrease. The curves show that the intergranular coupling between the grains, the critical current density and the flux pinning energy of the samples are increased by increasing the amount of cadmium up to $x = 0.04$. When the amount of cadmium is more than 0.04, the junctions between the grains are destroyed (see Fig. 5, sample $x = 0.06$). So, the intergranular

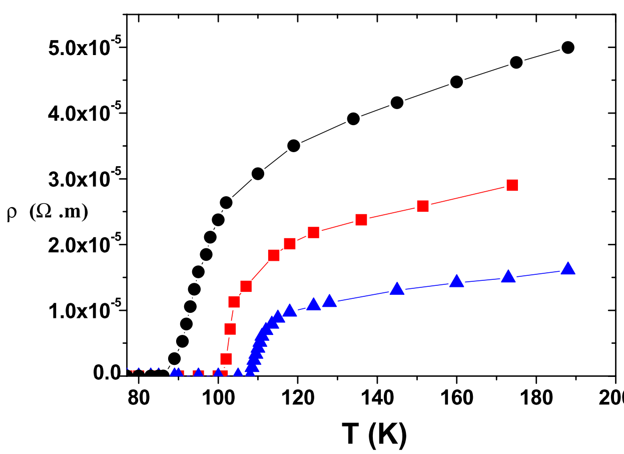

Fig. 2 ρ–T curves for different amounts of cadmium, 0.0 (■), 0.04 (▲) and 0.06 (●), and an annealing time of 270 h

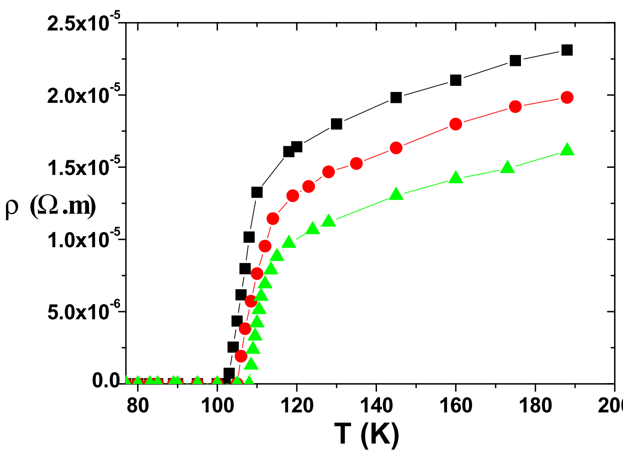

Fig. 3 ρ–T curves for different annealing times: 90 (■), 180 (●) and 270 h (▲) for the sample with a Cd content of 0.04

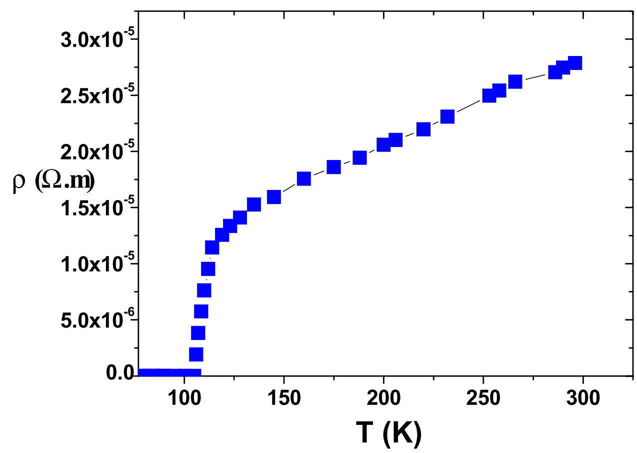

Fig. 4 The resistivity vs. temperature curve for the sample with a Cd content of 0.04 and an annealing time of 180 h

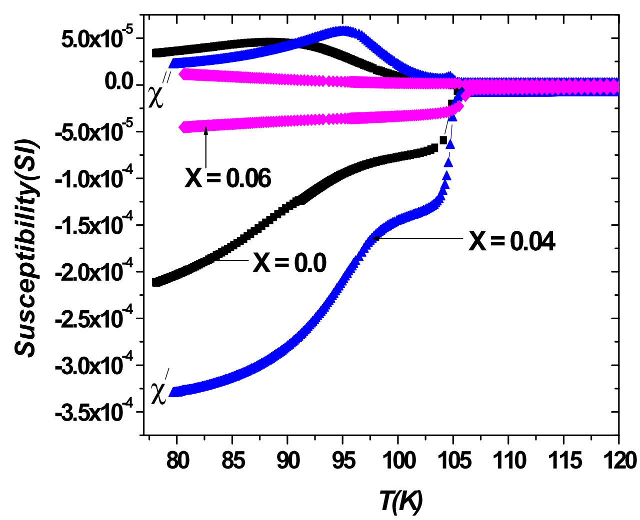

Fig. 5 Temperature dependence of magnetic susceptibility for samples with cadmium contents of 0.0, 0.04 and 0.06 and an annealing time of 270 h

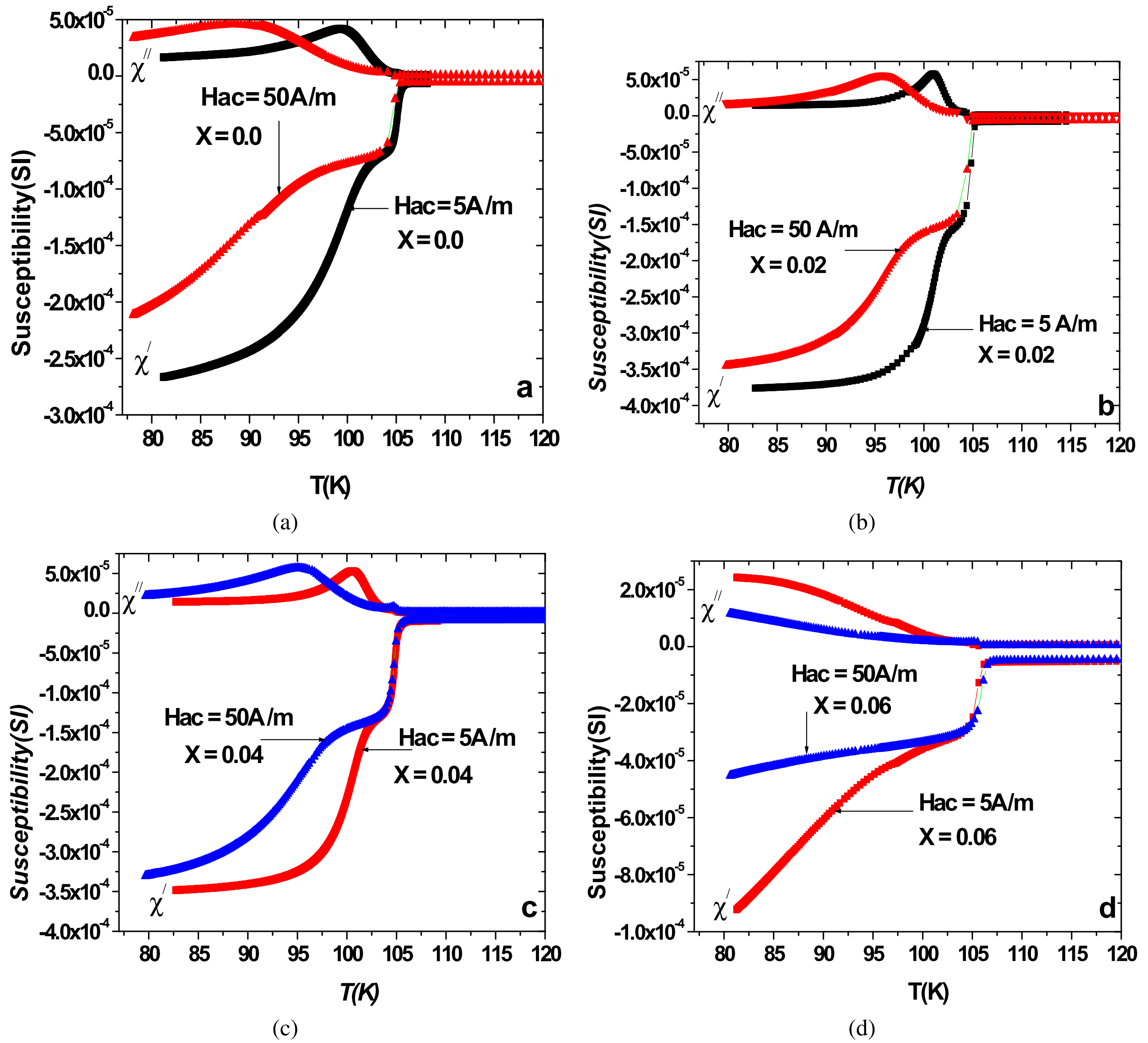

Fig. 6 Temperature dependence of magnetic susceptibility with fields 5 and 50 A/m for samples with cadmium contents of 0.0 (**a**), 0.02 (**b**), 0.04 (**c**) and 0.06 (**d**) and an annealing time of 270 h

coupling between the grains, the critical current density and the flux pinning energy in the sample with $x = 0.04$ are better than the sample with $x = 0.0$ and the sample with $x = 0.0$ is much better than the sample with $x = 0.06$.

Figure 6a–d show the results of AC magnetic susceptibility measurements for samples with $x = 0.0$, 0.02, 0.04 and 0.06 respectively and with an annealing time of 270 h for various AC fields. The diamagnetic onset temperature of the intrinsic superconducting transition for these samples is at about 107.5 K. It is clear from Fig. 6 that as the field increases the peak of χ'' shifts to lower temperatures and broadens. Also, the effect of the field on the intergranular component in the real part (χ') shifts to lower temperatures. The amount of shift is a function of the field amplitude, H_{AC}, which is proportional to the strength of the pinning force. When the pinning force becomes smaller, it shows that the critical current will decrease. The effect of the AC magnetic field on the sample with $x = 0.04$ is less than the other samples.

Moreover, for all the samples, the relation between the current and voltage in the region between J_{co} (onset of resistive state) and J_c (onset of normal state) follows the power law model $V = \alpha I^{\beta}$ [22, 23]: one of the sample curves is shown in Fig. 7. These results have been derived from the curves $\log V$–$\log I$ (see Figs. 8 and 9), which are plotted in the region between J_{co} and J_c. The value of β for each sample is given in Table 2. Considering the value of β and the relationship $\beta = U/k_B T$ [24], where U is the pinning

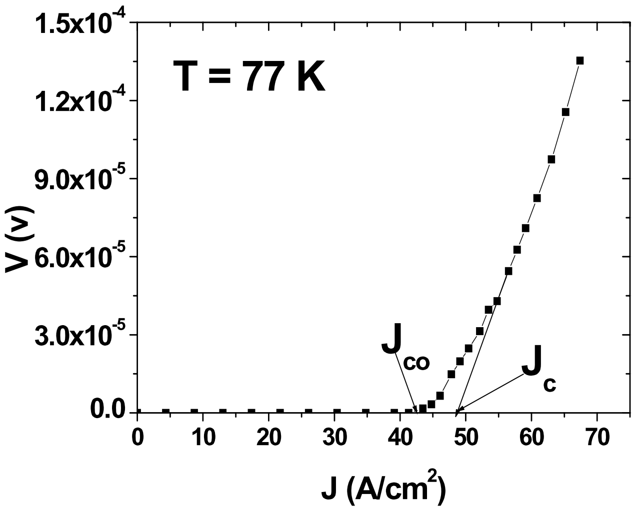

Fig. 7 V–J curves for the sample with a Cd content of 0.04 and an annealing time of 270 h

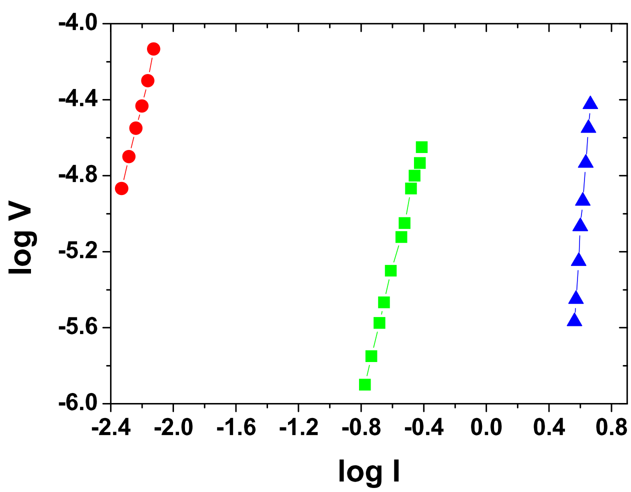

Fig. 8 log V–log I curves for samples with Cd contents: 0.02 (■), 0.04 (▲) and 0.06 (●) and an annealing time of 270 h

potential, it can be concluded that prolongation of the annealing time leads to an increase in flux pinning energy. In other words, the average force on the flux for depinning will increase.

Figure 10 shows the measured U vs. the amount of Cd in the samples for different annealing times. Fig. 10 shows that if the Cd content is increased, U will also increase. Also, on increasing the annealing time up to 270 h, U will increase as well. Hence, the maximum U was found for $Bi_{1.6}Pb_{0.36}Cd_{0.04}Sr_2Ca_2Cu_3O_y$ annealed for 270 h. When the amount of cadmium is more than 0.04, the amounts of unwanted phases, such as Bi-2212 and Bi-2201, will increase in the sample. These unwanted phases play the role of weak links [25, 26] and decrease the flux pinning energy.

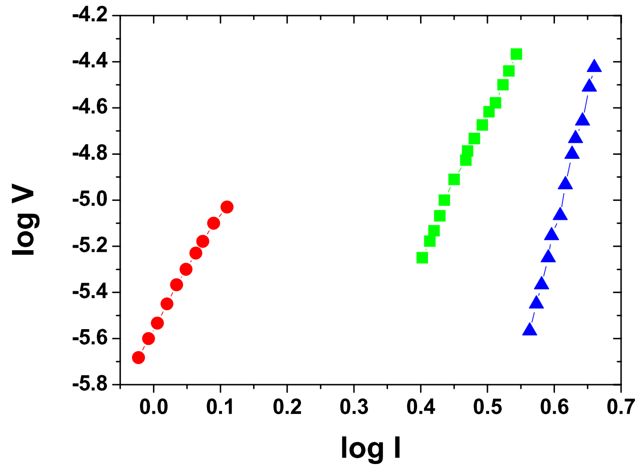

Fig. 9 log V–log I curves for different annealing times: 90 (■), 180 (●) and 270 h (▲) for the sample with a Cd content of 0.04

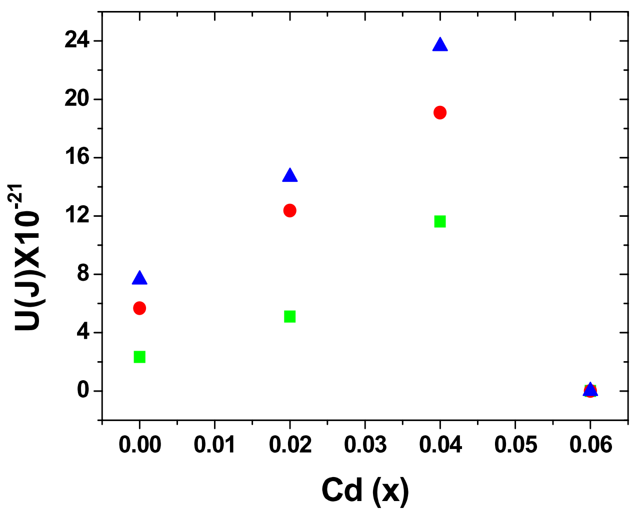

Fig. 10 The flux pinning energy of doped samples as a function of the cadmium amount for annealing times: 90 (■), 180 (●) and 270 h (▲)

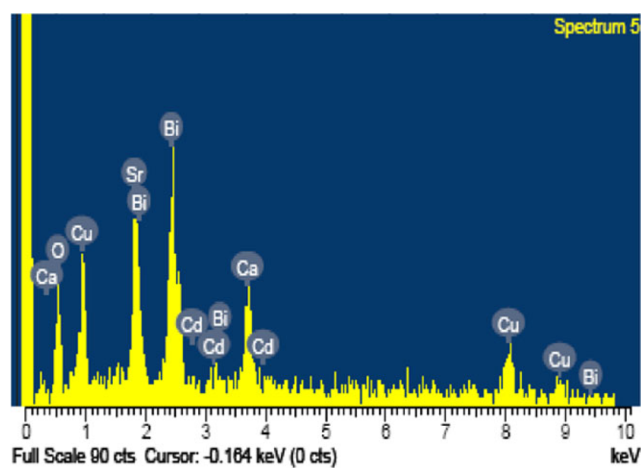

Fig. 11 EDX image of the sample with the cadmium amount of 0.04 and an annealing time of 270 h

Table 2 The results of measurement of $Bi_{1.64-x}Pb_{0.36}Cd_xSr_2Ca_2Cu_3O_y$ superconductor

x	β	T_c (offset) (K)	t (h)	U (J) $\times 10^{+21}$
0.0	2.2	92	90	2.33
0.0	2.35	97	180	5.67
0.0	7.21	101.3	270	7.64
0.02	4.81	95	90	5.103
0.02	11.67	99.3	180	12.37
0.02	13.85	102.3	270	14.68
0.04	10.96	102	90	11.62
0.04	18	105	180	19.08
0.04	22.30	107.5	270	23.64
0.06	0.02	77	90	0.0
0.06	0.06	83.5	180	0.0
0.06	0.21	86.2	270	0.0

The result of EDX measurements show that there is no unwanted element in the samples, which means that the samples are not contaminated during the synthesis process (see Fig. 11).

4 Conclusion

We have investigated the role of Cd in the synthesis of $Bi_{1.64-x}Pb_{0.36}Cd_xSr_2Ca_2Cu_3O_y$ with $x = 0.0$, 0.02, 0.04 and 0.06. The samples were prepared by a solid state reaction method using different annealing times. The results of AC magnetic susceptibility measurements for the samples show that the diamagnetic fraction of the sample with $x = 0.04$ is greater than the other samples. From the results, it can be concluded that the sample doped with cadmium having $x = 0.04$ and an annealing time of 270 h has an increase in percentage of the Bi-2223 phase. The effect of the AC magnetic field on the sample with $x = 0.04$ is less than the other samples. Our results also show that the maximum values of the flux pinning energy and T_c (offset) were obtained for the sample with an annealing time of 270 h and a Cd content of 0.04.

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